

# Fuel Staging for NO<sub>x</sub> Reduction in Biomass Combustion: Experiments and Modeling

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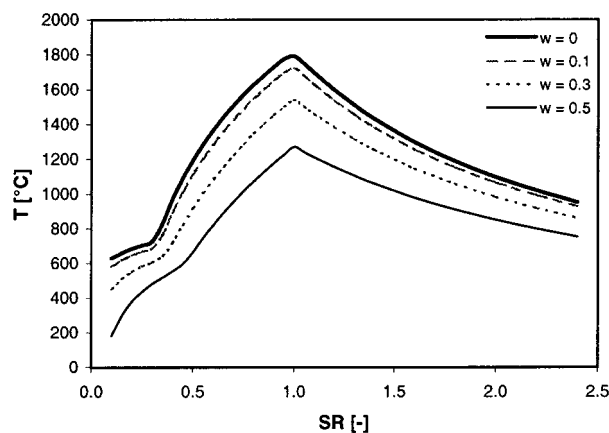
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With increasing use of biomass in combustion processes, the reduction of the related NO<sub>x</sub> emissions which originate mainly from the fuel nitrogen becomes more and more important. Efficient primary measures for NO<sub>x</sub> reduction are staged combustion techniques. Air staging has been investigated earlier and has found its way into practice. Since fuel staging has not been applied with nonpulverized biomass yet, the aim of the present work was to investigate the potential of fuel staging for NO<sub>x</sub> reduction in fixed bed systems. For this purpose, a prototype understoker furnace of 75 kW thermal input with two fuel beds in series was developed. Experiments were performed with wood chips (low nitrogen content) and UF-chipboards (high nitrogen content) to investigate the influences of the main process parameters, i.e., stoichiometric ratio, temperatures, residence time, and fuel properties on the conversion of fuel nitrogen to N-species. The most important parameters were found to be the temperature and the stoichiometric ratio in the reburn zone. The potential of fuel staging was measured and compared with air staging and unstaged combustion. The experiments show that low NO<sub>x</sub> emissions are already achievable with fuel staging at lower temperatures than with air staging, i.e., 900–1000 °C, and at a stoichiometric ratio of 0.85 in the reduction zone. The NO<sub>x</sub> reduction achieved under optimum conditions for UF-chipboard as main fuel was 78% which is higher than with air staging, where 72% NO<sub>x</sub> reduction was measured. For wood chips both measures attained about 66%. The nitrogen conversion during air and fuel staging has also been simulated using a furnace model based on ideal flow patterns as perfectly stirred reactors and plug flow reactors. A detailed reaction mechanism including the nitrogen chemistry (GRI-Mech 2.11) was implemented. The trends found with this model are in good agreement with the experiments and they indicate that even higher NO<sub>x</sub> reduction may be reached with improved process design. The investigations show that fuel staging is a promising technology for NO<sub>x</sub> reduction also for fixed bed biomass furnaces.

## Introduction

Biomass combustion systems emit nitrogen oxides which should be reduced since they contribute to the formation of acid rain and photochemical smog. The NO<sub>x</sub> emissions from biomass combustion are mainly caused by the nitrogen in the fuel.<sup>1</sup> Thermal NO<sub>x</sub> formation usually produces negligible amounts of NO<sub>x</sub> because the lower heating values leads to lower combustion temperatures compared to fossil fuels. Figure 1 shows the calculated adiabatic flame temperature of wood combustion as a function of the excess air ratio and the fuel water content. In Figure 2, the contributions of the different NO<sub>x</sub> formation mechanisms to the NO<sub>x</sub> emission as a function of temperature are displayed for fixed bed biomass combustion systems.<sup>2</sup> For any NO<sub>x</sub> formation mechanism, the oxygen content in the flame region and the residence time are also important parameters.



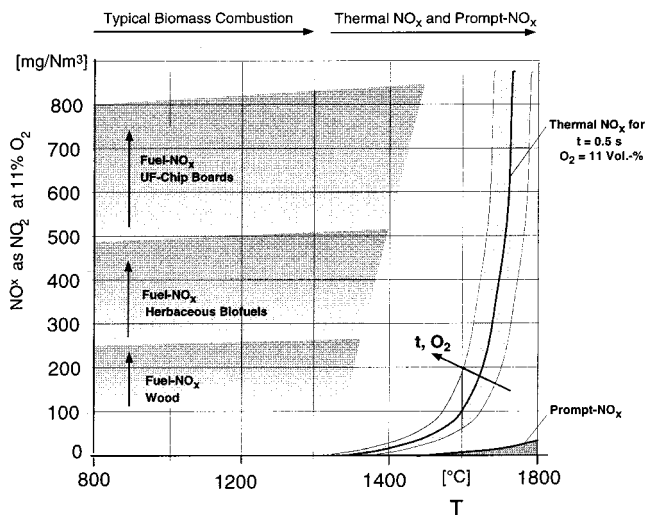
**Figure 1.** Influence of water content ( $w$ ) on the calculated adiabatic flame temperature of wood combustion with ambient air as function of the stoichiometric ratio SR (total pressure 1 bar, dry wood as CH<sub>0.7</sub>O<sub>1.4</sub>).

During recent years, primary measures have been developed to reduce the formation of nitrogen oxides during the combustion process. Staged combustion

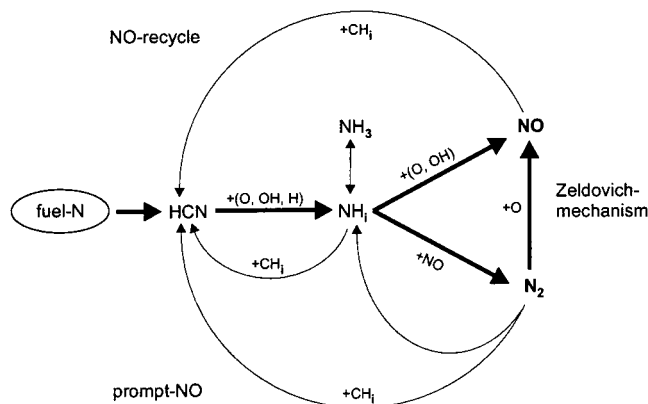
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(1) Nussbaumer, T. Schadstoffbildung bei der Verbrennung von Holz. Ph.D. Thesis ETH No. 8838, 1989.



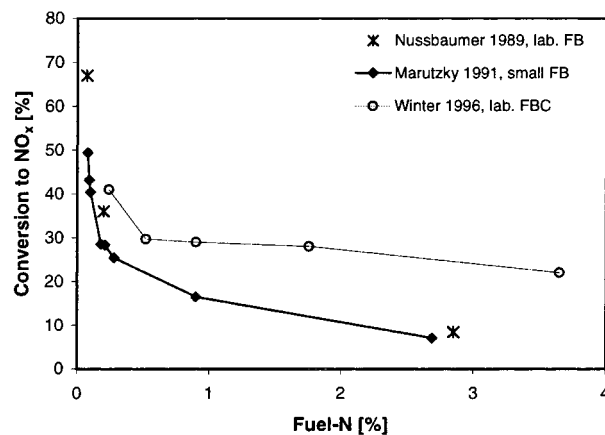
**Figure 2.** Influence of temperature on  $\text{NO}_x$  emission for biomass combustion.<sup>2</sup>



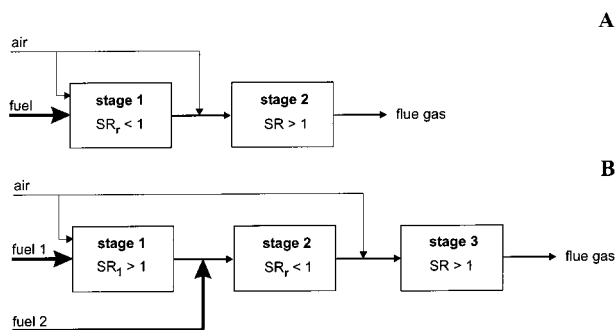
**Figure 3.** Simplified reaction path diagram of the  $\text{NO}_x$  formation and destruction in the gas phase.

techniques have been investigated for large scale power plants mainly, fired with gas, oil, or pulverized coal. Since the importance of renewable energy sources will grow in the future, clean combustion of solid biomass is important. But the fuel characteristics of biomass differ from fossil fuels, hence it remains to show which primary measures for low  $\text{NO}_x$  combustion can be applied for nonpulverized biomass.

During the thermal degradation of the biomass substance the main products containing fuel nitrogen after this conversion process are HCN and  $\text{NH}_3$ .<sup>3</sup> Further reactions then lead to molecular  $\text{N}_2$  or to NO when they are oxidized by the reaction with radicals (O, H, OH). Figure 3 shows a simplified global reaction path of fuel nitrogen. Only part of the total nitrogen is converted to  $\text{NO}_x$  as Figure 4 illustrates.<sup>1,4,5</sup> The goal of staged



**Figure 4.** Influence of fuel-N content on the conversion to N-species in combustion systems<sup>1,4,5</sup> (FB: fixed bed; FBC: fluidized bed combustor).



**Figure 5.** Principles of air staging (A) and fuel staging (B). combustion techniques is the promotion of the second reaction path by the creation of optimized conditions during combustion.

The reduction of fuel-N to molecular nitrogen in air staging is favored in the fuel rich primary combustion zone (Figure 5a). Investigations on fixed bed wood furnaces have shown that a minimum of the total fixed nitrogen ( $\text{TFN} = \text{HCN} + \text{NH}_3 + \text{NO} + \text{NO}_2 + 2\text{N}_2\text{O}$ ) emission from the primary combustion zone is reached for a stoichiometric ratio of 0.7 to 0.8 and a temperature of 1100–1200 °C and providing a mean residence time of 0.5 s.<sup>6</sup> After the reduction zone the combustion is completed in the burnout zone by injection of the excess air.

Air staging has high demands concerning the fuel properties. Because of the relatively high temperatures needed in the reduction zone the fusibility of the fuel ash is an important fuel property because ash sintering and slagging disturb the combustion process and therefore limits fixed bed combustion. Native wood usually has the highest melting range, about 1300 °C. The melting point of other wood fuels as UF-chipboard or demolition wood as well as of other biomass can be much lower, depending on their composition or whether they are mixed with other components. Herbaceous fuels for example have melting points at 800–900 °C because they contain relatively high amounts of various inorganic substances which can form new compositions with low melting points.<sup>7,8</sup>

Fuel staging or reburning has first been tested on coal fired utilities using natural gas as reburn fuel.<sup>9</sup> In fuel

(2) Nussbaumer, T. Primary and secondary measures for  $\text{NO}_x$  reduction in Biomass combustion. In *Developments in Thermochemical Biomass Conversion*; Blackie Academic and Professional: London, U.K., 1997.

(3) Aho, M.; Hämäläinen, J.; Tummavuori, J. Importance of solid fuel properties to nitrogen oxide formation through HCN and  $\text{NH}_3$  in small particle combustion. *Combust. Flame* **1993**, *95*, 22–30.

(4) Marutzky, R. Erkenntnisse zur Schadstoffbildung bei der Verbrennung von Holz und Spanplatten. *WKI-Bericht*; Fraunhofer Arbeitsgruppe für Holzforschung 1991, 26.

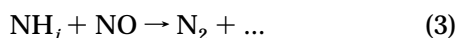
(5) Winter, F.; Wartha, C.; Hofbauer, H. A  $\text{NO}/\text{N}_2\text{O}$  classification system of single fuel particles. In *Developments in Thermochemical Biomass Conversion*; Blackie Academic and Professional: London, U.K., 1997; pp 1303–1315.

(6) Keller, R. Primärmaßnahmen zur  $\text{NO}_x$  Minderung. Ph.D. Thesis ETH No. 10514, 1994.

staging the first stage of the combustion process operates slightly fuel lean, whereby the NO<sub>x</sub> production is high (Figure 5b). Then, additional fuel is added which creates fuel-rich conditions in the reburn zone. When hydrocarbons are used as reburn fuel, the hydrocarbon radicals entering the reburn zone can initialize the NO reduction mechanisms. The major reaction path found from research studying the reburning mechanism through elementary chemical reactions under fuel rich conditions is the formation of HCN.<sup>10</sup>



Because the volatile content of wood is high (80–85%), wood is highly reactive and therefore well suited as reburn fuel. Also the nitrogen content may be beneficial since it leads to additional reducing species.<sup>11,12</sup>

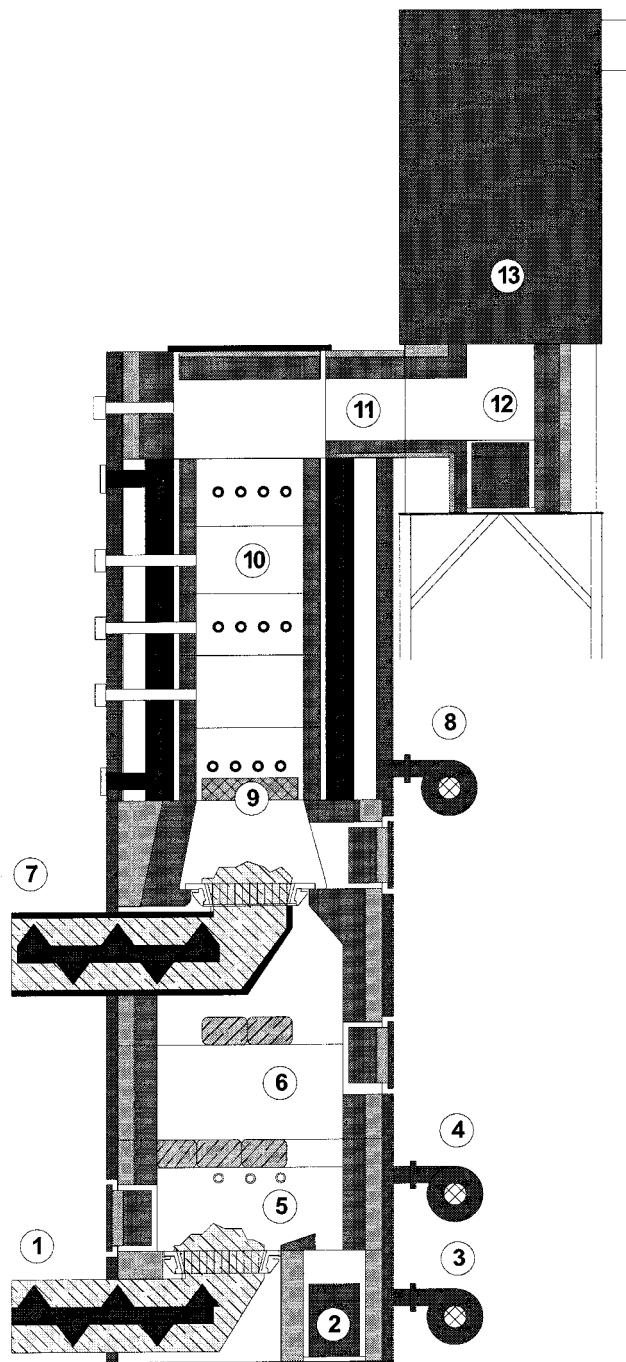


Further improvement of the reburning in power plants lead to the advanced reburning technologies where fuel staging is combined with other NO<sub>x</sub> reduction technologies.<sup>13</sup>

The aim of the work presented is to investigate whether a new concept for fuel staging based on fixed bed systems is suitable for the NO<sub>x</sub> reduction of non-pulverized wood fuels.<sup>14</sup> Focus of the research is the elaboration of the optimum combustion conditions for an efficient NO<sub>x</sub> reduction. The measured potential is compared with air staging. All the experiments presented are measured on a pilot-scale research facility of about 75 kW thermal input. To investigate the effect of the fuel nitrogen, wood chips (low nitrogen content) and UF-chipboard (high nitrogen content) are chosen for the experiments. The parameters investigated are the temperatures, the stoichiometric ratio, and the residence time in the reburn zone and the ratio of primary fuel and reburn fuel. The research facility also allows air staging conditions as well as unstaged combustion for comparison.

### Experimental Section

A new research facility was designed to investigate both air staging and fuel staging in a fixed bed furnace shown in Figure 6. The main components of the facility are two understoker



**Figure 6.** Research facility at the ETH Zurich (1. understoker (stage 1); 2. ash box; 3. and 4. air inlet; 5. primary zone; 6. burnout zone; 7. understoker (stage 2: reburn stage); 8. air inlet (level variable); 9. mixing element; 10. reduction zone; 11. burnout zone; 12. flyash box; 13. boiler).

grates in series. The alignment chosen for the furnace tries to approximate a straight plug flow of the gases, preventing dead zones and backflow regions.

The first stage is a conventional understoker furnace equipped with an air supply for primary and secondary air. Then, a small burnout chamber for the combustion gas follows. The design of the second grate is adapted to the higher gas flux due to the temperatures of 800–1200 °C. The maximum fuel input of both understoker systems is equal.

The second understoker is protected by a water cooled shell. To minimize the heat loss of the gas to the cooled surface the understoker housing is sheltered with bricks. Above the second grate, the reburn or reduction zone follows. At the entrance a

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**Table 1. Gas Analysis Equipment for the Measurements on the Research Facility**

location	species	method
flue gas	O <sub>2</sub>	paramagnetic
	NO, NO <sub>2</sub>	CLD
	CO, CO <sub>2</sub>	NDIR
reburn zone	C <sub>x</sub> H <sub>y</sub>	FID
	CO, CO <sub>2</sub>	NDIR
	CH <sub>4</sub>	NDIR
	H <sub>2</sub>	HCD
	O <sub>2</sub>	paramagnetic
	NO, NO <sub>2</sub>	CLD
	HCN, NH <sub>3</sub> , N <sub>2</sub> O	FTIR

static mixing element provides an enhanced mixing of the gases before passing the reburn zone. The reburn zone element has also a water cooled shell which keeps the reburn zone at low temperatures. The heat transfer can be influenced by the composition of the insulation layers. The reburn section has three levels of nozzles for the injection of the burnout air. According to the injection levels, the mean residence times can be varied: depending on the fuel inputs and temperatures from about 0.4 to 2 s. The reburn section also has several openings at different locations for gas sampling and temperature measurements. On the top, the burnout segment leads the gases further to the boiler unit of the facility. The lower part of the boiler unit still is designed as a combustion chamber. The upper part is the heat exchanger where the flue gas is cooled to 200–250 °C.

The furnace has no automatic ash removal. The ash of the first stage is collected in an ash box. The ash of the second stage has to be removed manually after 8 to 12 h of operation. A part of the flyash is collected in a box at the last redirection of the gas before entering the main heat exchanger.

Besides the fuel and the ash handling, the operation of the research facility is completely automated, controlled by a PLC. The first fuel stage is equipped with an oxygen control to keep the oxygen content within a narrow range ( $SR_1 = 1.1$  to 1.3). For fuel staging, it is essential that the stoichiometric ratio of the first stage is well controlled to enable understoichiometric conditions in the reduction zone with little reburn fuel input. The amount of reburn fuel, namely, is dosed according to the desired stoichiometric ratio in the reburn zone. The injection of burnout air completes the combustion after the reduction zone. The amount of burnout air is controlled the way that complete burnout is achieved at a stoichiometric ratio between 1.5 and 2.0. The oxygen content in the flue gas is also continuously measured with a second gas sensor. A flue gas ventilator controls the pressure in the facility and guarantees safe operation.

A more practical parameter than the stoichiometric ratio in the reburn zone is the reburn fuel rate RFR, which is the input of reburn fuel to the total fuel input.

$$RFR = m_2 LHV_2 / (m_1 LHV_1 + m_2 LHV_2) \quad (4)$$

where  $m_1$ ,  $m_2$  are the mass flows and  $LHV_1$ ,  $LHV_2$  are the heating values of the fuels of stages one and two. When the stoichiometric ratio of the first stage is known, the stoichiometric ratio in the reburn zone can be calculated as a function of RFR.

The measurements were done at stationary or quasi stationary conditions. The measurements of air flows, gas temperatures, gas composition, and heat output were monitored online. The gas composition was analyzed in the flue gas right after the boiler exit with industrial gas analyzers. For the analysis of the hot gas in the reduction zone a suction pyrometer combined with a probe for O<sub>2</sub> detection was used. With this probe also short fluctuations could be monitored with almost no delay time. Besides, a hot gas sampling line with different analyzers for measuring the gas in the reburn zone was installed. Table 1 gives an overview over the gas analysis equipment.

**Table 2. Fuel Properties of Wood Chips and UF-chipboard**

			wood chips	UF-chipboard
water		g/100 g	17.9	4.7
ash	wf <sup>a</sup>	g/100 g	1.0	1.3
volatiles	wf	g/100 g	83.0	79.9
char	wf	g/100 g	16.0	18.8
HHV		MJ/kg	16.13	18.9
LHV		MJ/kg	14.74	17.54
C	wf	g/100 g	49.6	49.2
H	wf	g/100 g	5.3	6.0
O	wf	g/100 g	44.9	41.1
N	wf	g/100 g	0.18	3.58
S	wf	g/100 g	0.03	0.12
NO <sub>2</sub> max from N at 11% O <sub>2</sub>		mg/Nm <sup>3</sup>	720	12300
bed ash properties:				
initial deformation temperature			1280 °C	1230 °C
hemispherical temperature			1310 °C	1260 °C
fluid temperature			1360 °C	1290 °C

<sup>a</sup> wf: water free base.

The experiments were carried out with wood chips and UF-chipboard (UF = urea formaldehyde) representing two biofuels with different nitrogen content. UF-chipboard has a very high nitrogen content compared to native wood because most of the nitrogen originates from the glue that holds the matrix of the chipboard. The wood chips were cut into small pieces of about 2–5 cm length by an ordinary wood hacker. The fine chipboard chips were pressed to briquettes with a diameter of about 8 cm. But the briquettes were crushed on the way to the grate by the transport system (screw feeders). Table 2 provides information about the fuel properties of the wood chips and the UF-chipboard used.

## Modeling

The aim of the modeling presented is the investigation of the influence of stoichiometry and temperature in the reburn zone on the NO<sub>x</sub> reduction by simulation of the fuel staging experiments with a model that needs moderate computational effort. Focus is placed on the homogeneous chemistry during reburning and influence of the operation conditions on the nitrogen oxides and their precursors. To keep the model as simple as possible, concessions on the modeling effort for the flow field are necessary.

The chemical reacting system is described as a combination of ideal reactor types such as the plug flow reactor (PFR) and the mixed flow reactor (PSR). The reactors in question are the reburn and burnout section of the fuel staged combustion. The resulting furnace model used with all the possible variants for simulation is shown in Figure 7. The input of the model are the combustion products of the first stage (gaseous only) and the reburn fuel. The combustion process after the reburn fuel input is separated and modeled as plug flow or mixed flow or both.

An important and critical process of the simulation is the calculation of the adequate inputs for the reburn zone modeling. Since all of the reburn fuel is converted in a fixed bed into gaseous species, the conversion of the solid reburn fuel is evaluated in a separate step, assuming complete fuel conversion.

At first, the main components of the combustion gas of the fuel-lean first fuel stage are calculated (O<sub>2</sub>, N<sub>2</sub>, CO<sub>2</sub>, H<sub>2</sub>O) by evaluating the combustion equation for

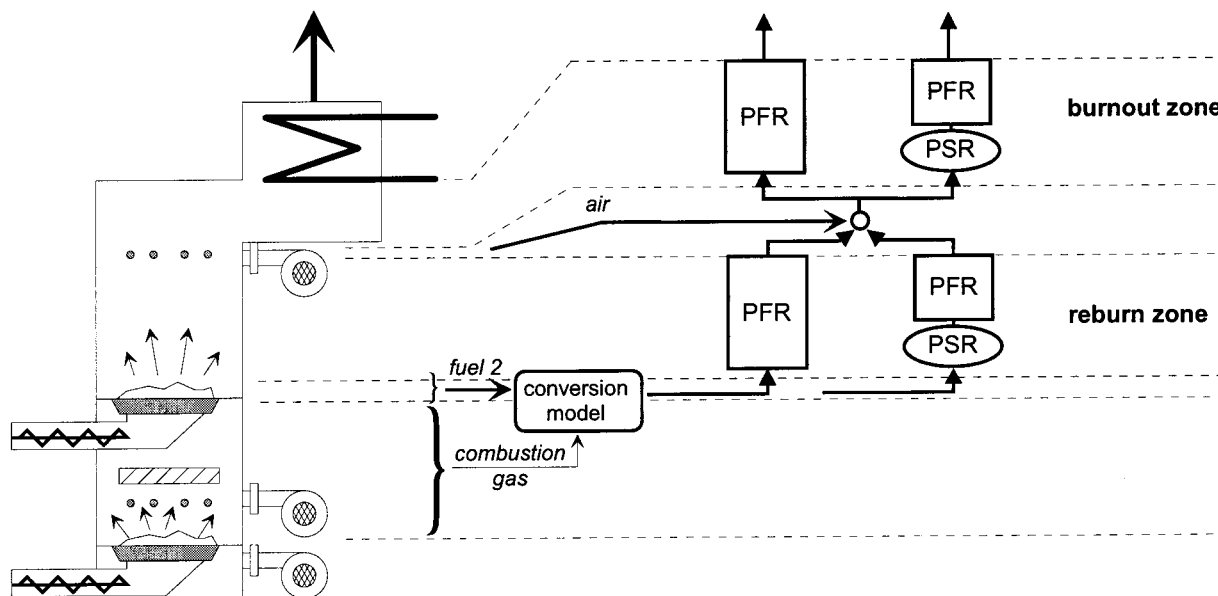


Figure 7. Scheme of furnace model for simulation.

Table 3. Conversion of Fuel Nitrogen into HCN and NH<sub>3</sub> Assumed for Simulation

	wood chips	UF-chipboard
O/N	223	11
conversion to HCN	0.03	0.007
conversion to NH <sub>3</sub>	0.05	0.005

wood as C<sub>x</sub>H<sub>y</sub>O<sub>z</sub> at a given stoichiometric ratio. Since the combustion gas of the first stage also contains NO<sub>x</sub> that effectively should be reduced, a specific amount of NO<sub>x</sub> is added to the calculated composition. The amount is derived from the measurements on the research facility. For UF-chipboard as main fuel a primary NO<sub>x</sub> emission of 385 ppm at SR<sub>1</sub> = 1.2 is assumed which represents the measurements. Afterward, the calculated gas of the fuel-lean first stage is used in the next step as donor of oxidizing agents for the gasification of the reburn fuel.

The detailed model for the thermal degradation of biomass into low molecular components is out of the scope of the present work and reported elsewhere.<sup>15</sup> The model for the conversion of the fuel-bound nitrogen during pyrolysis into HCN and NH<sub>3</sub> is based on experimental data of single particle pyrolysis at 800 °C.<sup>16</sup> Since the correlation found between the HCN/NH<sub>3</sub> ratio and the fuel-O/fuel-N ratio is valid for various fuels and nitrogen-containing model substances, it is also base for the implemented model. Thus, the pyrolysis gas composition of the reburn fuel is calculated next, assuming the conversion rates at a temperature of 800 °C. The specific amounts of HCN and NH<sub>3</sub> corresponding to this conversion are noted in Table 3.

For the fuel conversion under fuel rich conditions a model assuming chemical equilibrium is used for the calculation of the main components CO<sub>2</sub>, H<sub>2</sub>O, CO, H<sub>2</sub>, and CH<sub>4</sub> including C as solid phase.<sup>17</sup> Under adiabatic conditions at 800 °C, not all of the reburn fuel is converted into gaseous products at pyrolysis state as

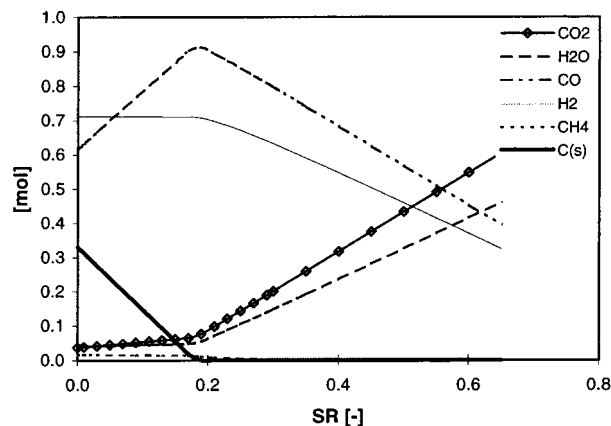


Figure 8. Calculated equilibrium composition for gasification of 1 mole of wood with ambient air (1 bar, dry wood as CH<sub>0.7</sub>O<sub>1.4</sub>) at 800 °C.

Figure 8 shows. An amount of char depending on the water content of the fuel and the process temperature remains. In a real furnace in fact, the charcoal burns out completely, also in the reburn stage since char burnout takes place after the pyrolysis. According to this fact, the amount of oxygen needed for the char burnout is computed and subtracted from the amount of O<sub>2</sub> available in the gas coming from the first fuel stage. Then, the pyrolysis gas and the combustion gas are mixed proportionally to the given reburn fuel rate.

The main advance of the chosen model is the possibility of handling detailed reaction mechanisms for investigating the gas-phase reactions. There are several reaction mechanism for natural gas (methane) combustion including nitrogen chemistry available, including more or less species and elementary reactions. The mechanism selected for the present model is the GRI-Mechanism version 2.11 (49 species, 279 reactions).<sup>18</sup>

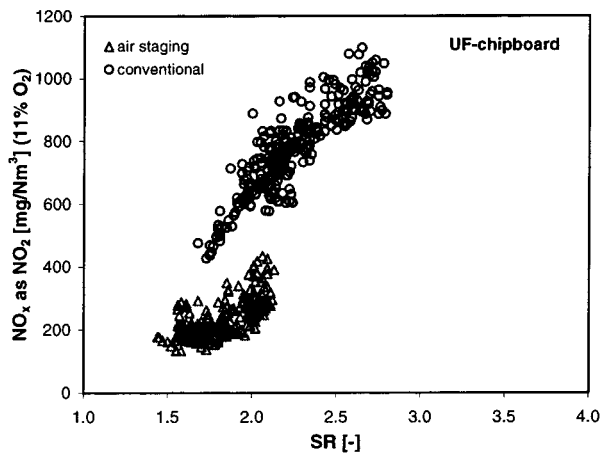
For modeling the gas-phase reactions of the furnace described model, the CHEMKIN II software package

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(16) Hämäläinen, J. Effect of fuel composition on the conversion of fuel-N to nitrogen oxides in the combustion of small single particles. Ph.D. Thesis, University of Jyväskylä, 1995.

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(18) [http://me.berkeley.edu/gri\\_mech/](http://me.berkeley.edu/gri_mech/).



**Figure 9.**  $\text{NO}_x$  emissions as a function of stoichiometric ratio for conventional and air staged combustion of UF-chipboard.

was used.<sup>19</sup> The calculation of the input and output data of the different processes is accomplished with separate input routines.

The aim of the model simulations is to predict the influence of the process parameters on the nitrogen species as a group, and not the calculation of absolute emission values. Therefore, a reference value named reduction rate  $R$  is introduced, allowing a dimensionless presentation of the results. The definition of  $R$  is

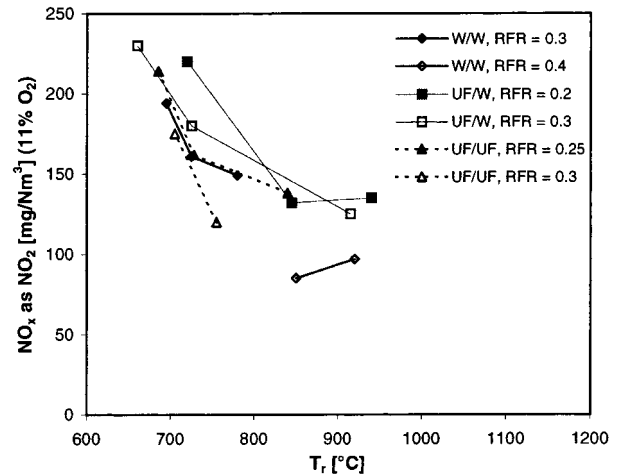
$$R = 1 - (\text{TFN}/\text{TFN}_0) \quad (5)$$

where  $\text{TFN}_0$  is the input of all nitrogen species to the reburn zone at the beginning of the simulations. Assuming that all nitrogen species besides  $\text{NO}$  and  $\text{NO}_2$  emitted from the first fuel stage are negligible, this value corresponds to the measured  $\text{NO}_x$  emission of the first fuel stage.

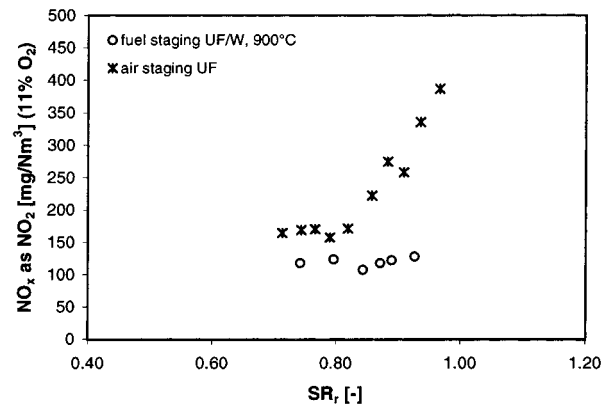
## Results and Discussion

The experiments with air staging carried out on the research facility confirm the previously shown  $\text{NO}_x$  reduction potential. The experiments prove that the stoichiometric ratio in the reduction zone has a distinct influence on the  $\text{NO}_x$  emission of the combustion process, providing that the optimum conditions for temperature, mean residence time, and mixing rate are also accomplished. The experiments indicate the important role of the fuel rich zone created by air staging. Figure 9 opposes air staging and conventional combustion and shows the  $\text{NO}_x$  reduction potential of air staging. Correlating the  $\text{NO}_x$  emissions of air staging to the corresponding total stoichiometric ratio demonstrates the reduction potential of about 72% for UF-chipboard, meaning from 550 to 150  $\text{mg}/\text{Nm}^3$   $\text{NO}_x$  at 11%  $\text{O}_2$ . For wood chips, the reduction measured is about 65%, corresponding to a  $\text{NO}_x$  reduction from 260 to 90  $\text{mg}/\text{Nm}^3$ .

A summary of the experimental results for fuel staging with different fuel combinations is presented in Figure 10. The measured  $\text{NO}_x$  emissions are plotted as a function of the temperature in the reburn zone for



**Figure 10.** Influence of temperature in reburn zone on  $\text{NO}_x$  emission for different RFR (UF = UF-chipboard; W = wood chips;  $\text{CO} < 500 \text{ mg}/\text{Nm}^3$  at 11%  $\text{O}_2$  for all data,  $\text{SR}_1 = 1.2$  to 1.3).

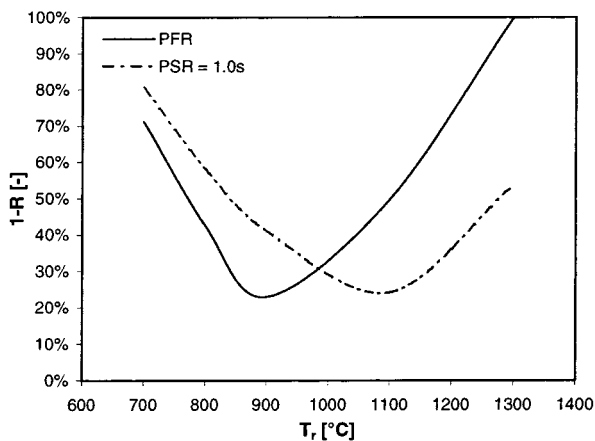


**Figure 11.** Influence of the stoichiometric ratio in the reduction zone on the  $\text{NO}_x$  emissions for air staging (UF-chipboard:  $T$  in reduction zone at 1050–1080 °C,  $\text{CO} < 250 \text{ mg}/\text{Nm}^3$  at 11%  $\text{O}_2$ ) and fuel staging (UF-chipboard main, wood chips reburn fuel:  $T$  in reduction zone at 900 °C,  $\text{SR}_1 = 1.25$ ,  $\text{CO} < 300 \text{ mg}/\text{Nm}^3$  at 11%  $\text{O}_2$ ).

different reburn fuel rates. The  $\text{CO}$  emissions of the evaluated measurements were at least below 500  $\text{mg}/\text{Nm}^3$  at 11%  $\text{O}_2$ , but usually below 250  $\text{mg}/\text{Nm}^3$ . Although only a few settings were measured, some trends are visible. The measurements clearly demonstrate that the  $\text{NO}_x$  emission can be reduced significantly with reburning. Around 750 °C the  $\text{NO}_x$  emissions already begin to decrease. Minimum emissions for UF-chipboard of 120  $\text{mg}/\text{Nm}^3$  are reached, whereas for wood chip combustion less than 100  $\text{mg}/\text{Nm}^3$  at a temperature between 800 and 900 °C are measured. The emission level for wood chip combustion is therefore comparable to the one measured with air staging. The  $\text{NO}_x$  emissions reached for the combustion of UF-chipboard are about 20 to 25% lower than with air staging as Figure 11 shows. The figure also indicates that the optimum stoichiometric ratio in the reduction zone is slightly higher for fuel staging than for air staging, meaning about 0.85 compared to 0.8.

The optimum temperature range for  $\text{NO}_x$  reduction begins above 850 °C for all fuel combinations tested. But above 700 °C already the  $\text{NO}_x$  reduction begins to be effective. A decrease of the reduction potential with increasing temperature cannot be seen within the

(19) Kee, R.; Rupley, F.; Miller, J. Chemkin II: Fortran chemical kinetics package for the analysis of gas-phase chemical kinetics. SAND89-8009 1990, Sandia National Laboratories.



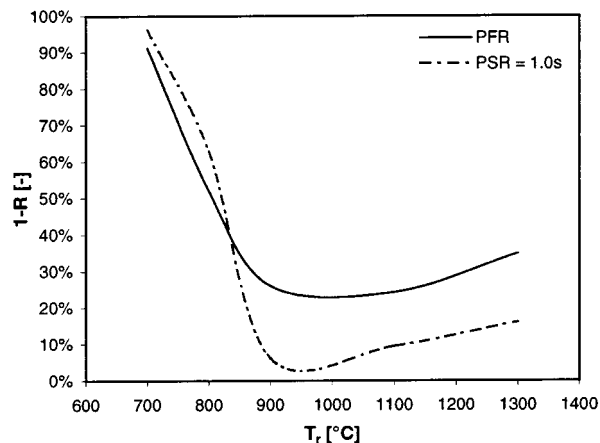
**Figure 12.** Influence of temperature on calculated reduction rate after reburn zone for RFR = 0.25 ( $\approx 0.92$ ) for UF/UF (UF-chipboard main and reburn fuel; total mean residence time in reburn zone = 2 s).

measured data range, although the data of fuel staging with wood chips at wood chip combustion (*W/W*) indicate a minimum at about 800 °C. However, concerning the temperatures presented it has to be mentioned that the gas temperature measured with the suction pyrometer are up to 100 °C higher than the temperature indicated by simple thermocouples which are the base for the data.

The residence time of the gas in the reduction zone is calculated with the approximated gas flow and the given geometry. The maximum residence time is  $\tau$  (air nozzles at position 3), which corresponds to a mean residence time of about 2 s for most of the experiments. If the air is injected through the nozzles at position 2, the resulting mean residence time is shortened to about  $(5/9)\tau$ . The measurements indicate little influence of the residence time, i.e., the NO<sub>x</sub> emission at  $(5/9)\tau$  is about 10% higher compared to the maximum residence time.

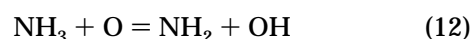
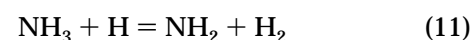
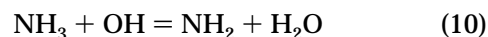
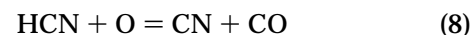
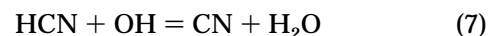
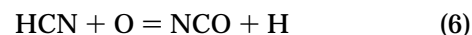
The simulation results show the calculated influence of the reburn temperature and the reburn fuel rate on the reduction rate  $R$  for UF-chipboard as main and reburn fuel. To simulate the influence of the mixing conditions, once a simple plug flow (PFR) is imposed and once a combination of a mixed flow (PSR) and a PFR with different mean residence times, the total mean residence time for the reburn zone is fixed for all cases to 2 s.

First, the reburn zone of the fuel staging is simulated, using different models for the mixing mode which is modeled by a variable splitting of the mean residence times between the mixed reactor (PSR) and the plug flow reactor (PFR). The reaction mechanism used is the GRI-mechanism. Figure 12 and Figure 13 show the reduction rate for different RFR. Considering the curves for plug flow (PFR) only, their trends indicate a more or less wide optimal temperature range starting at about 850 °C and ending at 950 °C or 1100 °C, depending on RFR. With increasing RFR, the temperature range with high reduction becomes wider. When mixed flow is allowed, the trends are shifted toward higher temperatures, but simultaneously the reduction rates increase. The figures show that the choice of the mixing model parameters has a considerable influence on the results.



**Figure 13.** Influence of temperature on calculated reduction rate after reburn zone for RFR = 0.35 ( $\approx 0.79$ ) for UF/UF (UF-chipboard main and reburn fuel; total mean residence time in reburn zone = 2 s).

According to the GRI-mechanism, the decomposition of HCN and NH<sub>3</sub> follows the reactions:



This reaction path is valid over the entire temperature range investigated. Toward low temperatures, almost all NO is reduced, but the model predicts an increasing formation of N<sub>2</sub>O and NO<sub>2</sub>.

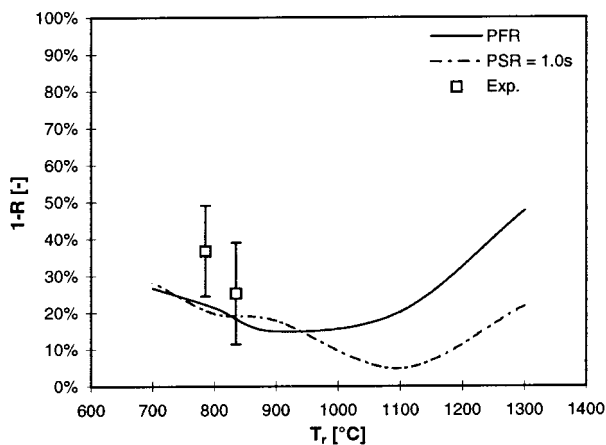
With increasing temperature, more and more HCN and NH<sub>3</sub> are decomposed, HCN in higher rates than NH<sub>3</sub>. The N<sub>2</sub>O formation is reduced drastically. The higher decrease of the reduction rate for RFR = 0.25 is due to a diminished NO reduction. The main cause is the recombination of HNO with H, OH, or O to NO. However, with increasing temperature at low RFR the reaction rates of NO forming and reducing reactions fall.

The temperature has also a distinct influence on the reaction velocities. Regarding the nitrogen species forming and consuming reactions, the characteristic reaction time at 800 °C is about 10 times slower than at 1300 °C.

The NO<sub>x</sub> emissions presented are measured in the flue gas, after the burnout zone of the test facility. Therefore, the measured emissions have to be compared with simulation results that include the modeling of the burnout stage. This means that the gas of the fuel rich reburn zone has to react with the burnout air to the flue gas. For the simulation, the input data for the burnout modeling are the output data of the reburn simulations.

Figure 14 shows an example of the calculated reduction rate after burnout assuming plug flow in the burnout zone (isothermal PFR at 1300 °C with 2 s mean





**Figure 14.** Calculated reduction rate after burnout zone for RFR = 0.30 ( $\approx 0.85$ ) for UF/UF (UF-chipboard main and reburn fuel; total mean residence time of burnout = 2 s).

residence time). The two curves in the figure represent the two different reburn models, once PFR only and once PSR plus PFR in series. The figure shows that the low reduction rates at lower temperatures calculated in the reduction zone before are transformed to higher reduction rates. This observation is true especially for the PFR curve. Toward higher temperatures the influence of the burnout on the reduction rate is negligible. The maximum reduction predicted varies between 82% and 95%, depending on the model for the reburn zone. However, the calculated reduction rates are higher than the measured.

The burnout conditions also have an important influence on the emission of the nitrogen species. The analysis of the simulations indicates that at high burnout temperature  $\text{NH}_3$  and HCN are destroyed leading only to a weak increase of the  $\text{NO}_x$  emission.  $\text{N}_2\text{O}$  which is formed at low temperatures in the reduction zone is also reduced to a negligible amount at this burnout temperature. The emission of  $\text{NO}_2$  is small compared to  $\text{NO}$ .

The observed change of the reduction rate at low temperatures may be influenced by the burnout modeling. A different temperature regime in the burnout zone for example may modify the reduction rates for low temperatures in the reburn zone. But calculations assuming isothermal plug flow at 1100 and 800 °C show that the burnout temperature has less influence on the overall reduction rate, providing that the preliminary reburn temperature is higher than 900 °C. Similar observations are reported from experiments on a laboratory plug flow reactor.<sup>20</sup>

The comparison of the measured data with the calculated  $\text{NO}_x$  reduction indicates that the theoretical potential of fuel staging is even higher than the experiments on the research facility have demonstrated. However, the trends generally show a good concordance. The lower reduction rates at low temperatures confirms the role of the temperature not only in the reburn zone, but also in the burnout zone. Additionally, the simulations point at the importance of the mixing and flow conditions within the furnace.

## Conclusions

The present investigation has demonstrated that fuel staging can be successfully applied for the combustion of nonpulverized wood fuels in fixed bed combustion. For UF-chipboard as reburn fuel, a  $\text{NO}_x$  reduction of up to 78% was achieved. The temperature range for an efficient reduction of the nitrogen species by homogeneous reactions is lower than for air staging. The experiments show that low  $\text{NO}_x$  emissions are already achievable at relatively low temperatures, meaning at 900–1000 °C. Therefore, fuel staging may be a favorable method for fuels with a high nitrogen content. Fuel staging is also favorable for fuels with high ash contents and low ash fusion temperature because it allows us to keep the combustion temperature of the first fuel stage below 1000 °C. The temperature control can be accomplished by heat extraction. As the temperature in the burnout zone after the reburn zone should be high enough to ensure complete burnout of the gas, a reasonable use of the transferred heat could also be the preheating of the burnout air.

All reburn fuels investigated showed a  $\text{NO}_x$  reduction potential at comparable optimum temperature and stoichiometric ratio. For high reduction rates, a mean residence time in the reduction zone of about 1.5 s regarding the examined facility is provided.

When a fixed bed is used for reburning with nonpulverized fuels, a high voidage of the fuel bed is beneficial to the reburning because a deep penetration of the combustion gas enhances and accelerates the fuel conversion rates and improves the mixing of the gas phase. For this reason, effects or conditions that make the passing of fuel bed for the gases more difficult, i.e., a dense fuel bed or slagging conditions, should be avoided. Concerning the design of the reburn zone, its shape should promote the gas mixing and prevent an inhomogeneous flow field.

Fuel staging demands some higher investment in the hardware and higher control of the furnace. On the other hand, costs for additional reducing agents and catalysts are saved, an important advantage over SCR and SNCR techniques. The reducing agent is the fuel itself. Additionally, a sophisticated management of the furnace switching between air staging and fuel staging may even result in advantages of the part load behavior compared to other  $\text{NO}_x$  reduction techniques.

The model which considers detailed chemical reaction kinetics predicts a higher  $\text{NO}_x$  reduction potential than the measured. The difference between model simulation and measurements indicates that the furnace design and the process control can be further improved. The influence of the reburn temperature and stoichiometry is well represented by the model.

However, the model is a suitable tool for the qualitative study of the influence of the main process parameters and allows a fast and easy simulation of the effect of each variation.

**Acknowledgment.** The research was funded by the Swiss Federal Office of Energy and the Swiss Federal Office for Education and Sciences. The technical support of Tiba Muller Ltd. is gratefully acknowledged.

(20) Kristensen, P.; Glarborg, P.; Dam-Johansen, K. Nitrogen chemistry during burnout in fuel staged combustion. *Combust. Flame* 1996, 107, 211–222.